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sulfur induced stable  
negative ion  
formation**

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# On the influence of fuel sulfur induced stable negative ion formation on the total concentration of ions emitted by an aircraft gas turbine engine: comparison of model and experiment

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Abstract

A model which considers the formation and evolution of combustion ions in a combustor of an aircraft engine in dependence on the electron detachment efficiency from negative ions is presented. It is a further development of the model reported by (Sorokin et al., 2003). The model allows to consider the effect of the transformation of primary negative ions to more stable secondary negative ions with a much higher electron affinity and as a consequence a greater stability with respect to electron thermal detachment. The formed stable negative ions most probably are sulfur-bearing ions. This effect slows down the charged particle neutralization rate leading to an increase of the concentration of positive and negative ions at the combustor exit. The results of the simulation and their comparison with the ground-based experimental data obtained within the framework of the project PartEmis (Particle emission, measurements and predictions of emission of aerosols and gaseous precursors from gas turbine engines; coordinator: C. Wilson) at the QinetiQ test facility at Pyestock, UK (Wilson et al., 2003) support the above hypothesis, i.e. the increase of the fuel sulfur content leads to an increase of the ion concentration at the combustor exit.

1. Introduction

Gaseous ions emitted by aircraft gas-turbine engines are of considerable interest as they may promote the formation of aerosols, contrails and perhaps even of clouds (Frenzel et al., 1994; Yu and Turco, 1998; Arnold et al., 1998 and 2000; Sorokin and Mirabel, 2001). Recently gaseous positive and negative ions have been measured in the exhaust of an aircraft engine just after the exit of the combustor in a test rig at the ground (Wilson et al., 2003; Haverkamp et al., 2003). These measurements prompted an interesting finding, namely an influence of the fuel sulfur content (FSC) on the total concentrations of positive ( $n_P$ ) and negative ( $n_N$ ) ions and this influence was found to vary with the fuel flow (FF) into the combustor. It was hypothesized by the authors of

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this work, that at high FSC sulfur-bearing negative ions rapidly form and these ions have a rather high stability with respect to electron thermal and associative electron detachment, even at the high temperatures encountered in the combustor. Thereby the concentration of free electrons decreases and that effect slows down the neutralisation rate of charged particles leading to an increase of  $n_P$  and  $n_N$ . The present paper investigates that hypothesis by a theoretical model which considers the formation of charged particles in the combustor. The results of the model calculations support the above hypothesis.

## 2. Model

The model used for the present investigation builds on the model recently developed by Sorokin et al. (2003), but has the following modifications: (i) transformation of the primary negative ions  $O_2^-$  to secondary negative ions  $X^-$  is introduced; (ii) all  $X^-$  ion species are lumped together and are assumed to experience electron detachment by a thermal or an associative mechanism which is slower than for  $O_2^-$ ; (iii) the rate of electron detachment from  $X^-$  relative to the rate of electron detachment from  $O_2^-$  is introduced explicitly as an adjustable parameter  $f_{det}$  in order to simulate the fuel sulphur-induced build-up of stable sulphur bearing negative ions.

For a high fuel sulfur content (HFSC) such as the one used in the PARTEMIS project experiments (Wilson et al., 2003) i.e. 1270 mg of S per kg of fuel (i.e. 1270 ppm), the expected concentration of  $SO_2$  molecules is about of  $1.6 \times 10^{15} \text{ cm}^{-3}$ . This leads to a sufficiently rapid conversion of  $O_2^-$  ions to  $SO_2^-$  ions via electron transfer from  $O_2^-$  to  $SO_2$  since the electron affinities (EA) of neutral  $O_2$  and  $SO_2$  molecules are 0.45 and 1.11 eV, respectively, and the rate coefficient is close to the upper collision rate coefficient, i.e.  $1 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$  at  $T = 1500\text{--}2000 \text{ K}$ . The mean  $O_2^-$  lifetime with respect to reaction with  $SO_2$  is hence around  $10^{-6} \text{ s}$ . In comparison the  $O_2^-$  lifetime with respect to thermal electron detachment is about of  $1 \times 10^{-8} \text{ s}$  for  $T = 1500\text{--}2000 \text{ K}$  (Möhler et

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al., 1992) and therefore electrons and  $O_2^-$  are in a quasi-equilibrium. In addition, in the exhaust flow, a substantial fraction of the  $O_2^-$  ions may experience conversion to the more stable sulfur ions. In fact the typical residence times of the combustion products in a post-flame zone (of about 5 ms) and in the post-combustor zone (of about 5–10 ms) is much longer than the time needed for these reactions to occur.

The  $SO_2^-$  may further react with  $SO_3$  via electron transfer as  $EA(SO_3)=2.06$  eV leading to  $SO_3^-$  which in turn rapidly reacts with  $SO_2$  and  $O_2$  resulting in  $SO_4^-$  and  $SO_5^-$  ion formation (Möhler et al., 1992). These ions are more stable with respect to electron detachment as they have a much larger electron bond energy ( $EA=5.1$  eV). Building on recent gaseous sulfur (VI) measurements (Katragkou et al., 2003) in the exhaust of an aircraft gas-turbine engine at the combustor exit, a concentration of  $SO_3$  of about  $(2-5)\times 10^{13} \text{ cm}^{-3}$  is expected. This leads to an  $SO_2^-$  lifetime of about  $5\times 10^{-5}$  s with respect to the reaction with  $SO_3$ . The lifetime of  $SO_3^-$  with respect to the reaction with  $O_2$  leading to the formation of  $SO_5^-$  is much shorter ( $\approx 10^{-9}$  s) because of the high concentration of oxygen molecules in the post-flame zone ( $O_2 \sim 5\times 10^{18} \text{ cm}^{-3}$ ). This last calculation is based on a rate constant  $k=2.5\times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  (Möhler et al., 1992) and a temperature dependence proportional to  $T^{-1/2}$ . Thus the build-up of negative ions which resist electron detachment much better than  $O_2^-$  is fast compared to the average residence time of combustion products in the post-flame zone of the combustor. The effect of a partial conversion of primary negative oxygen ions to secondary sulphur-bearing ions (such as  $SO_3^-$ ,  $SO_4^-$  or  $HSO_4^-$ ,  $SO_5^-$ ) will be important for the concentration of free electrons only if the concentration of these secondary ions is comparable with the “initial” concentration of combustion ions formed in the flame zone.

The concentration of primary combustion ions depends on their formation rate  $Q$  in the flame zone and particularly in the flame front (Fialkov, 1997; Sorokin et al., 2003) which in turn depends on the fuel flow (FF) in the combustor (Lefebvre, 1985). Therefore, it is reasonable to assume that  $Q$  directly depends on the fuel flow (FF) into the combustor. As a consequence for low FF (LFF), when the intensity of the formation

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of primary combustion ions is lower than for high FF, stable sulfur-ion formation may have an effect on the concentrations of free electrons and ions. More free electrons may lead to a higher rate of electron-ion recombination and hence to a lower exit concentration of combustion ions if, of course, there is no other more effective process of electron interaction (e.g. soot particles). By contrast for high FF (HFF), due to the very high concentration of primary negative ions, it is possible that the conversion of  $O_2^-$  ions to sulfur ions will have no effect on electrons and  $O_2^-$  concentrations. In this case, it is expected that the variation of FSC will not influence the total emission of chemiions. Unfortunately, there exist considerable uncertainties concerning the detailed mechanism of the evolution of negative ions in the combustor. Therefore, the present model does not attempt to treat in all details the complex interactions between neutral and charged species. Rather the model considers the relative efficiency  $f_{det}$  of the electron detachment process from all possible  $X^-$  ions as a free input parameter. This parameter which can be varied between 1 to 0 reflects two possible limiting cases (i)  $f_{det}=1$  means the absence of  $O_2^-$  conversion into secondary more stable negative ions (in this case, no effect of FSC on ion concentrations is expected), (ii)  $f_{det}=0$  means that  $O_2^-$  is fully converted into  $X^-$  ions. The aim of the present study is therefore to investigate the influence of  $f_{det}$  on the concentration of free electrons ( $n_e$ ), positive ( $n_p$ ) and negative ( $n_N$ ) ions in the combustor. The equations and reaction coefficients used are described in detail in a recent paper by Sorokin et al. (2003). The model consists of a system of coupled differential equations, which describe the time evolution of the concentrations of electrons, negative and positive ion and also multi-charged soot particles. These equations describe the following processes: the chemiionization rate  $Q(t)$  for the production of primary electrons and positive ions in the combustion zones; the soot-particle charging (multi-charged particles are allowed) due to both electron thermo-emission or electron and ion attachment to soot particles; the electron-ion and ion-ion recombination; electron attachment to and detachment from neutral molecules. Furthermore, we consider mono-dispersed soot particles ( $r_s=20$  nm), and the effect of soot oxidation in the combustion and dilution zones as well as soot-particle coagulation

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are neglected. For all calculations, the soot-particle concentration was assumed to be  $n_s = 2 \times 10^7 \text{ cm}^{-3}$ . The corresponding soot emission indexes  $EI_N = 2 \times 10^{14}$  particles and  $EI_S = 0.03 \text{ g soot per kg fuel}$  are consistent with the measured values at the aircraft engine exit (Petzold et al., 1999). Finally, the work function for electron thermal emission from soot surfaces was taken as 5 eV in agreement with recent measurements made on combustion carbon particles (Matter et al., 1995).

### 3. Results

Figures 1a–c show for three values of  $f_{det} = 0.001$ , 0.01 and 0.1 (neither  $f_{det} = 0$  or 1 will be achieved in a real case) and for  $Q = 2 \times 10^{14} \text{ cm}^{-3} \text{ s}^{-1}$  the concentration of free electrons  $n_e$ , positive  $n_p$  and negative ions  $n_N$  as a function of time in the combustor where the residence time is  $\tau_c = 5 \text{ ms}$ . It can be seen from these figures that  $n_e$  initially increases steeply reaching its maximum values at about  $t = 1 - 2 \mu\text{s}$ . After  $t = 2 \mu\text{s}$  the concentration of electrons decreases which reflects the attachment of electrons to oxygen molecules. The decrease of  $n_e$  is of course more pronounced for the smaller  $f_{det}$ , i.e. when for example  $\text{O}_2^-$  ions are fully converted into  $\text{X}^-$  ions with a high EA. Comparing Figs. 1a and 1c,  $n_e$  is seen to increase from  $5 \times 10^4$  to  $1.3 \times 10^6 \text{ cm}^{-3}$  at  $t = 5 \text{ ms}$  when  $f_{det}$  varies from  $10^{-3}$  to  $10^{-1}$ .

Negative-ion formation becomes appreciable after  $t = 1 \mu\text{s}$ . Their concentration then increases very steeply, due to the attachment of electrons to  $\text{O}_2$  molecules until it reaches a maximum at about 0.2 – 0.4 ms. The negative-ion concentrations  $n_N$  are seen to vary with  $f_{det}$ , i.e. they go down from  $2 \times 10^8$  to  $4 \times 10^7$  at  $t = 5 \text{ ms}$  when  $f_{det}$  increases from  $10^{-3}$  to  $10^{-1}$ , and this reflects the effect of electron detachment. The concentration of positive ions  $n_p$  in the combustor exhibits less variations with  $f_{det}$  but we still see a slight decrease of  $n_p$  when  $f_{det}$  varies from  $10^{-3}$  to  $10^{-1}$ . This reflects the fact that, as  $f_{det}$  increases, more free electrons are available for recombination with positive ions. Since the electron-ion recombination coefficient  $\alpha_{EP}$  exceeds markedly

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the ion-ion recombination coefficient  $\alpha_{ij}$  more positive ions are neutralized as  $f_{det}$  increases.

These figures show that  $n_N$  and  $n_P$  do not change much after  $t=20\ \mu\text{s}$ . This shows that ion losses by electron-ion or ion-ion recombination and ion attachment to soot particles are not very efficient after this time. For example, for  $n_N \sim n_P = 2 \times 10^8\ \text{cm}^{-3}$ ,  $\alpha_{PN} \sim 5 \times 10^{-8}\ \text{cm}^3/\text{s}$ , and  $T$  in the range 1500–2000 K, the time scale of the ion-ion recombination process is about of 0.5 s which markedly exceeds the time span ( $\tau_c = 0.005\ \text{s}$ ) of ions in the combustor. Finally, in Figs. 1a–c are indicated the experimentally determined ion concentrations at the exit of a combustor obtained during the PARTEMIS campaign. The indications HFSC and LFSC correspond to fuel sulfur contents of 1270 and 50 ppm, respectively. For HFF this concentration is close to  $2 \times 10^8\ \text{cm}^{-3}$ .

Figures 2a–b show the variation of the ratios  $n_N/n_{N0}$  and  $n_P/n_{P0}$  with  $f_{det}$  at the combustor exit ( $n_{N0}$  and  $n_{P0}$  are the concentrations corresponding to the fixed value  $f_{det} = 0.02$ , which represents the limit after which the curves merge) for three values of the primary chemiion formation rate  $Q = 10^{14}$ ,  $2 \times 10^{14}$  and  $10^{15}\ \text{cm}^{-3}\ \text{s}^{-1}$ . For the negative ions, one can see that as  $f_{det}$  increases,  $n_N/n_{N0}$  decreases significantly and that this decrease is more pronounced for smaller values of  $Q$ . By contrast with negative ions, positive-ion concentrations are hardly influenced by  $Q$  or  $f_{det}$ . If  $Q$  is sufficiently large ( $Q > 10^{14}\ \text{cm}^{-3}\ \text{s}^{-1}$ ) their evolution in the combustor depends mainly on ion-ion recombination. However, for smaller values of  $Q$  the concentration of negative ions is not large enough to lead to substantial depletion by ion-ion recombination and therefore the concentration of positive ions remains approximately constant in the combustor. In addition, for high enough values of  $Q$  ( $> 10^{14}\ \text{cm}^{-3}\ \text{s}^{-1}$ ) the exit concentrations of positive and negative ions are approximately equal (Sorokin et al., 2003).

Figure 3 shows the variation of the exit ratios  $n_P(f=0)/n_P(f=0.02)$  and  $n_N(f=0)/n_N(f=0.02)$  as a function of  $Q$ . For positive ions this ratio remains approximately constant in spite of the variation of  $Q$  over 6 orders of magnitude. The situation for negative ions is quite different. For small values of  $Q$  the decrease of the electron de-

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attachment rate (to the limit  $f_{det}=0$ ) leads to a marked increase of the exit concentration as compared to  $f_{det}=0.02$ . Such an influence of  $Q$  and  $f_{det}$  on the exit concentration of negative ions is directly connected to the predominance of the electron attachment processes leading to the formation of negative ions, over their depletion by ion-ion recombination. Smaller  $f_{det}$  lead to larger  $n_N$  behind the flame zone and larger exit  $n_N$ . Of course, increasing  $Q$  leads to a stronger influence of ion-ion recombination and hence to a weaker dependence of the exit concentrations on  $f_{det}$ . In Fig. 3 are also indicated the experimental values (Haverkamp et al., 2003) of the ratio  $(n_N)_{HFSC}/(n_N)_{LFSC}$  for the LFF and HFF cases. For HFF this ratio is close to unity while for LFF it is close to 1.5. It follows from Fig. 3 that there exists a small range of  $Q$  values ( $10^{14} - 10^{15}$ ) for which the model is consistent with the measurements. For larger  $Q$  there is no dependence on  $f_{det}$  and for smaller  $Q$  this dependence is too strong. Also for small values of  $Q$  the simulated exit concentrations (e.g.  $n_P=10^7$  and  $n_N=8 \times 10^7 \text{ cm}^{-3}$  for  $Q=10^{13} \text{ cm}^{-3} \text{ s}^{-1}$  and  $f_{det}=0$ ) are lower than the corresponding measurements ( $\approx 2 \times 10^8 \text{ cm}^{-3}$  for both positive and negative ions).

Figure 4 shows, for  $Q=2 \times 10^{14}$ , the variation of  $n_N$  and  $n_P$  with  $f_{det}$  at the combustor exit. It can be seen that both concentrations are only slightly affected by the variation of  $f_{det}$  (less than a factor of two) and that this variation is even less pronounced for positive ions. This behavior mostly reflects the stabilizing effect of the ion-ion recombination.

Thus the performed simulation shows that the model presented is able to predict experimental exit concentrations of positive and negative ions and to explain the observed trend in these concentrations with variation of FSC for different FF into the combustor. However, the model also predicts that the simulated result depends on some input parameters which are not well known (e.g.  $Q$  and  $f_{det}$ ). So together with the measurement of the ion exit concentrations, the measurements of other parameters are needed. For example, simulations show that for very low values of  $f_{det}$  ( $\approx 0$ ) the exit concentration of free electrons is very small ( $< 10^{-4} \text{ cm}^{-3}$ ) However, already for  $f_{det}=0.001$  the exit electron concentration is  $n_e \geq 10^4 \text{ cm}^{-3}$ . The later value probably can be measured. Also, the simulation yielded the expected result that the exit fractions of positively and

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negatively charged soot particles are in some dependence on the parameters  $Q$  and  $f_{det}$  and also depend on electron and ion concentrations. So it would be desirable to further investigate the interaction between electrons and ions and soot particles by simultaneous measurements of both ion and charged soot-particle exit concentrations.

- 5 However, in the given model, we consider only large soot particles and do not take into account the possible formation of charged and soot particles directly via the chemiionization mechanism. In general, the charging of smallest primary soot particles should influence the soot-particle aggregation process due to a change of the effective coagulation rate coefficient.

#### 10 4. Conclusions

A numerical model has been developed to consider the effect of the formation of stable negative ions with high electron bond energy on the concentration of positive and negative ions at the combustor exit of an aircraft gas-turbine engine. The model includes the formation of primary electrons and positive ions by chemiionization, electron attachment and detachment to neutral gas species, the ion-ion recombination, the interaction of electrons and ions, and the electron thermo-emission from the hot soot surfaces and the removal of charged species by soot particles in the combustor. Results of the simulation are compared with data obtained by MPI-K Heidelberg in the framework of the PartEmiss project.

- 20 The comparison has shown that the formation of secondary stable negative ions such as  $\text{SO}_x^-$  ( $x=3-5$ ) and as a consequence the induced decrease of the overall electron detachment coefficient, may lead to an increase of the concentration of ions at the combustor exit. This effect is most pronounced for a not too high chemiionization rate in the flame zone of the combustor. Both these results are confirmed by observations, i.e. the increase of the measured concentration of ions at the exit of the combustor with increasing FSC (fuel sulfur content) for low fuel flow into the combustor and a comparatively essentially smaller effect for high fuel flow. The performed simulation

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and comparison with measurements have shown that the emission of combustion ions really may strongly depend on the sulfur chemistry and sulfur-bearing ion formation. This process should be also important for the charging of mainly smallest primary soot particles in the combustor.

- 5 *Acknowledgements.* Support of this work by the European Community through the contract G4RD-CT-2000-00207 (PARTEMIS) and by the INTAS project OPEN 2000-460 and by the Max Planck Society is gratefully acknowledged.

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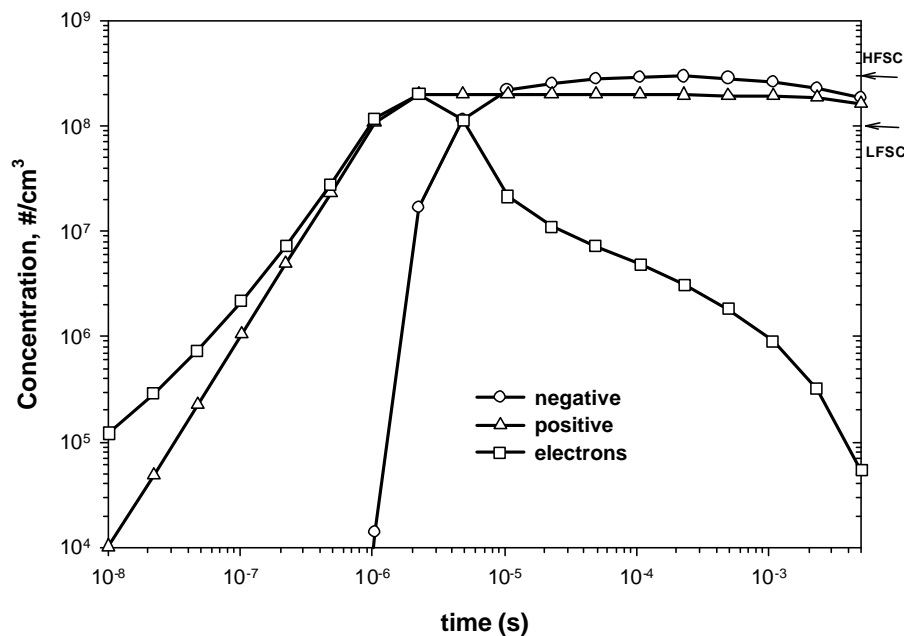
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**Fig. 1.** Concentration profiles of electrons, positive and negative ions in the combustor for different values of the electron detachment efficiency  $f_{det}=0.001$  (a), 0.01 (b) and 0.1 (c). Also shown for comparison are the experimental data.

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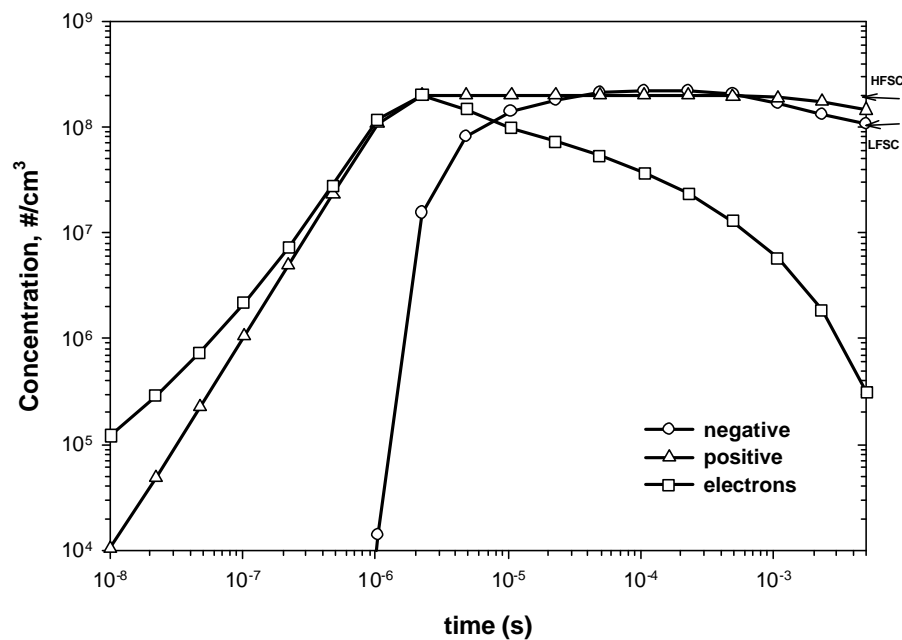


Fig. 1. (b) Continued.

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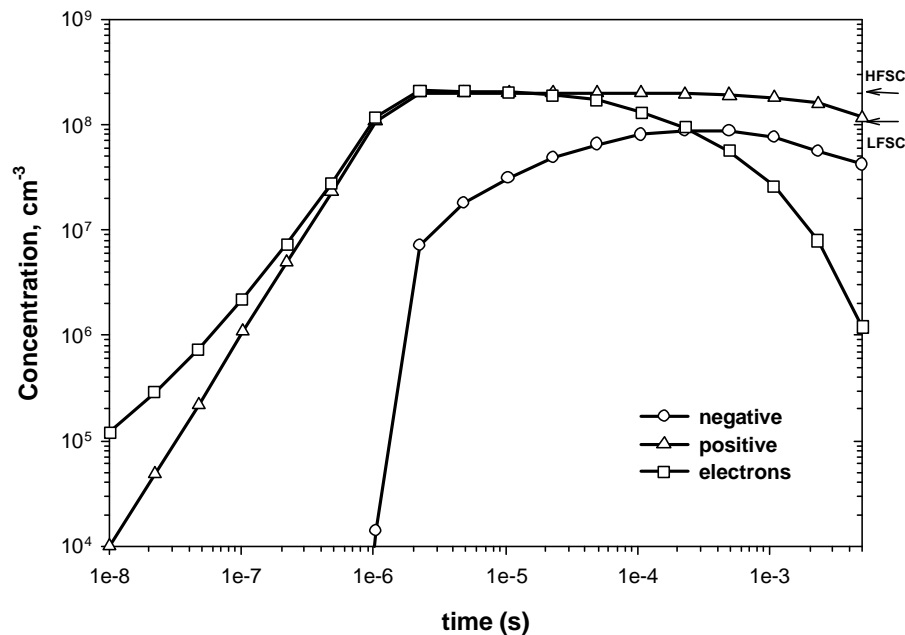
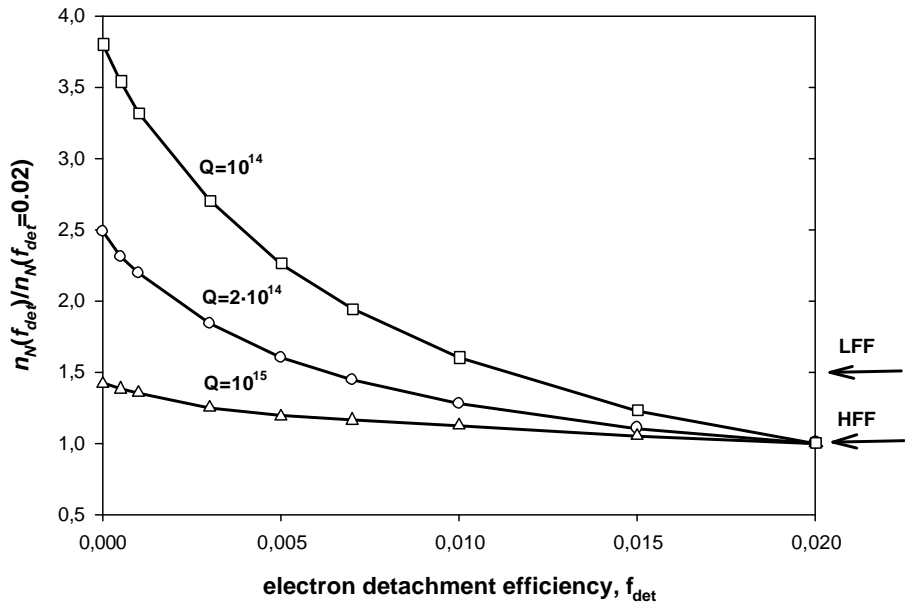


Fig. 1. (c) Continued.

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**Fig. 2.** Relative concentration of negative (a) and positive (b) ions at the combustor exit versus the electron detachment efficiency for different values of the chemiion formation rate  $Q$  in the flame zone. Also shown for comparison are the experimental data.

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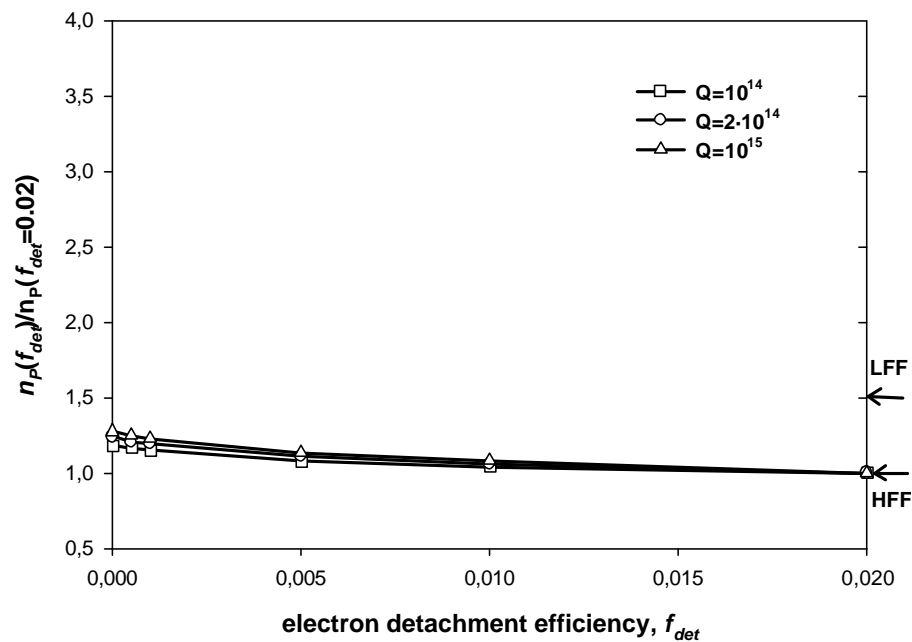
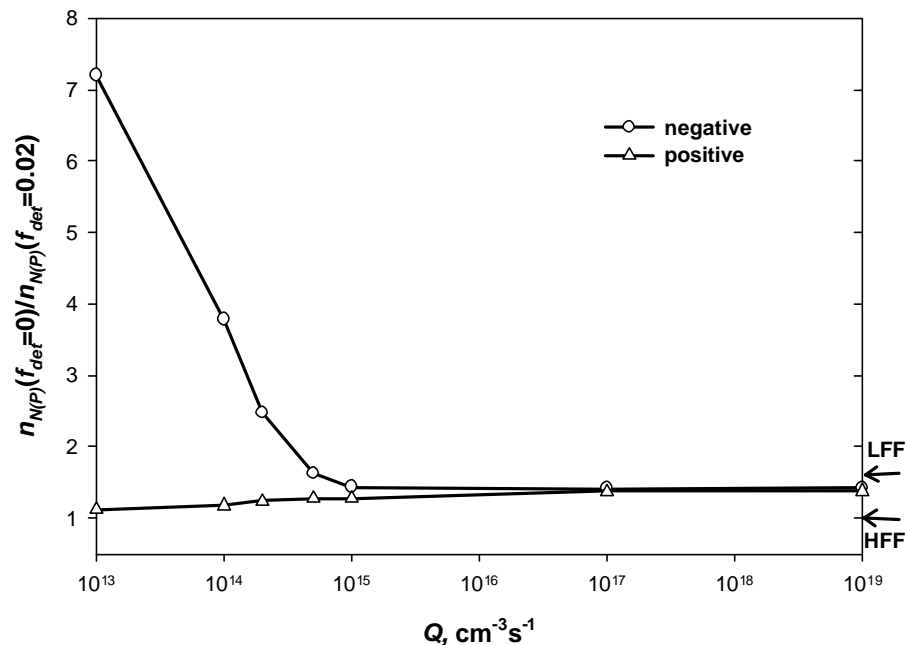


Fig. 2. (b) Continued.

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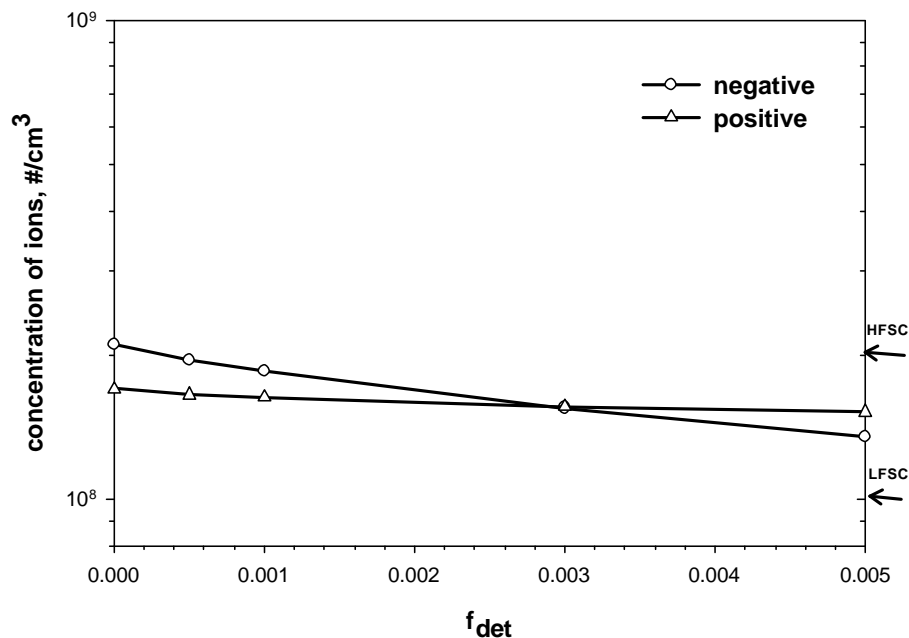


**Fig. 3.** Ratio of the concentrations of positive and negative ions at the combustor exit for two values of the electron detachment coefficient  $f_{det}=0$  and 0.02 versus the chemion formation rate  $Q$ . Also shown for comparison are the experimental data.

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**Fig. 4.** Concentration of positive and negative ions at the combustor exit versus the electron detachment efficiency for  $Q=2 \times 10^{14} \text{ cm}^{-3} \text{ s}^{-1}$ . Also shown for comparison are the experimental data.

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